

JC14 Rec'd PCT/PTO 17 JAN 2002



19036.3

U.S. APPLICATION NO

10/031232

**TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US).  
CONCERNING A FILING UNDER 35 U.S.C. 371**

INTERNATIONAL APPLICATION NO.  
PCT/EP00/06881

INTERNATIONAL FILING DATE  
July 19, 2000

PRIORITY DATE CLAIMED  
July 23, 1999

TITLE OF INVENTION: Method for Producing a Composite Material, Composite Material Produced According to Said Method, Molded Part That Consists of Such a Composite Material and Method for the Production Thereof

APPLICANT(S) FOR DO/EO/US

ZIEGLER, Maik et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(I)
4. ☒ A proper Demand for International Preliminary Examination was made by the 19<sup>th</sup> month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C.371(c)(2)).
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C.371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C.371(c)(3)).
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C.371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C.371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C.371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included
13. ☒ A FIRST preliminary amendment.  
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
  1. Form PCT/IB/308
  2. Return Postcard
  3. Conditional Petition to Revive

17. The following fees are submitted:				CALCULATIONS PTO USC Only	
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1)-(5)):					
Search Report has been prepared by the EPO or JPO..... \$ 890.00					
International preliminary examination fee paid to USPTO.....\$710.00					
No international preliminary examination fee paid to USPTO but international search fee paid to USPTO.....\$740.00					
Neither international preliminary examination fee nor international search fee paid to USPTO.....\$1040.00					
International preliminary examination fee paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4).....\$100.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$	890
Surcharge of \$130.00 for furnishing the oath or declaration later than ____20 ____30 months from the earliest claimed priority date (37 CFR 1.492(e))				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	51 - 20 =	31	X \$ 18.00	\$	558
Independent claims	1 - 3 =	0	X \$ 84.00	\$	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$ 280	\$	
TOTAL OF ABOVE CALCULATIONS =				\$	1448
Reduction by 1/2 for filing by small entity, if applicable.					
SUBTOTAL =				\$	1448
Processing fee of \$130.00 for furnishing the English translation later than ____ 20 ____30 months from the earliest claimed priority date + 130					
TOTAL NATIONAL FEE =				\$	1448
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	40
TOTAL FEES ENCLOSED =				\$	1488
				Amount to be:	
				refunded	\$
				charged	\$

a. \_\_\_\_ A check in the amount of \$ \_\_\_\_\_ to cover the above fees is enclosed.

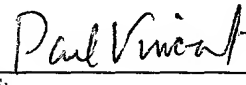
b. X Please charge my Deposit Account No. 50-0698 in the amount of \$ 1488 to cover the above fees.

c. X The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-0698. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be filed and granted to restore the application to pending status.

Please send all correspondence  
by AIRMAIL  
to:

Dr. Paul J. Vincent  
Lichti, Lempert & Lasch  
Bergwaldstr. 1  
D-76227 Karlsruhe  
  
Fed.Rep. of Germany



SIGNATURE:

Dr. Paul J. Vincent

NAME

37,461

REGISTRATION NUMBER

10/031232

531 Rec'd PCT

17 JAN 2002

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: ZIEGLER, Maik et al. ) Examiner:  
PCT Application No.: PCT/EP00/06881 ) unknown  
PCT Filing Date: July 19, 2000 ) Art Unit:  
 ) unknown

For: METHOD FOR PRODUCING A COMPOSITE MATERIAL, COMPOSITE  
MATERIAL PRODUCED ACCORDING TO SAID METHOD, MOLDED PART  
THAT CONSISTS OF SUCH A COMPOSITE MATERIAL AND METHOD  
FOR THE PRODUCTION THEREOF

Docket No.: 19036.3

Assistant Commissioner for Patents  
Washington, DC 20231  
U.S.A.

PRELIMINARY AMENDMENT

Please enter this amendment prior to calculation of the  
filing fees. This amendment is based on the translation of  
the application as filed on July 19, 2000.

IN THE SPECIFICATION:

On page 1, insert as a title prior to the first paragraph --  
BACKGROUND OF THE INVENTION --.

On page 4, insert as a title prior to the third paragraph --  
SUMMARY OF THE INVENTION --.

On page 20 insert as a title prior to the brief description  
of the drawings --

BRIEF DESCRIPTION OF THE DRAWING --.

On page 20 insert as a title following the brief description  
of the drawing --

DESCRIPTION OF THE PREFERRED EMBODIMENT --.

On page 22, line 1, replace as a title "Claims" with --  
WE CLAIM: --.

IN THE CLAIMS:

Please delete claims 1 - 51 without prejudice and enter new  
claims 52 - 102 as indicated below.

52. A method for producing a composite material, the method  
comprising the steps of:

- 10034222-01702
- a) selecting a particulate foam of thermoplastic material having a low crystallite portion in an otherwise amorphous phase, said particulate foam consisting essentially of one of polyalkylene terephthalate and a blend of polyalkylene terephthalates;
  - b) disposing a layer of material proximate to said particulate foam;
  - c) heating said particulate foam to a temperature in a region of a particulate foam melting temperature to melt together said particulate foam for forming a molded body;
  - d) connecting together said particulate foam and said layer of material during or following step c); and
  - e) tempering, during step c), during step d), or following steps c) and d), said particulate foam at a temperature converting said amorphous phase into an increased overall crystallite portion.
53. The method of claim 52, wherein step c) comprises the steps of forming said molded body using pressurized hot vapor and controlling at least one of a pressure and a pressurizing time for tempering said particulate foam.
54. The method of claim 52, further comprising additionally tempering said molded body following step c).

55. The method of claim 52, further comprising slowly cooling said molded body following step c).
56. The method of claim 52, further comprising controlling a temperature during tempering using a DSC measurement.
57. A composite material, consisting essentially of a molded body of particulate foam and at least one layer connected to said molded body produced in accordance with the method of claim 52, wherein said particulate foam consists essentially of one of a polyalkylene terephthalate and a blend of polyalkylene terephthalates.
58. The composite material of claim 57, wherein said particulate foam consists essentially of polyethylene terephthalate (PET).
59. The composite material of claim 57, wherein said particulate foam further comprises at least one of synthetic and natural reinforcing fibers.
60. The composite material of claim 57, wherein said layer connected to said particulate foam is a cover layer forming a visible side of the composite material.

61. The composite material of claim 60, wherein said cover layer is a foil comprising at least one thermoplastic polymer.
62. The composite material of claim 60, wherein said cover layer has a textile structure.
63. The composite material of claim 60, wherein said cover layer comprises at least one of polyalkylene terephthalate, PET, and a blend of polyalkylene terephthalates.
64. The composite material of claim 60, wherein said cover layer is welded to said particulate foam.
65. The composite material of claim 60, wherein said cover layer is coated onto said particulate foam.
66. The composite material of claim 57, further comprising an intermediate layer of fibers disposed between said cover layer and said particulate foam.
67. The composite material of claim 66, wherein said intermediate layer comprises at least one of a fiber mat, cloth, tissue, knitted fabric, and knits.

68. The composite material of claim 67, wherein fibers of said intermediate layer consist essentially of at least one of polyalkylene terephthalate, PET, and a polyalkylene terephthalate blend.
69. The composite of claim 66, wherein said intermediate layer comprises at least one of synthetic and natural reinforcing fibers.
70. The composite material of claim 66, wherein said intermediate layer is welded to said particulate foam.
71. The composite material of claim 66, wherein said intermediate layer is welded to said layer of material.
72. The composite material of claim 66, wherein said the layer of material is laminated onto said intermediate layer.
73. The composite material of claim 66, wherein said intermediate layer is a mixed fiber layer containing a first portion of at least one of polyalkylene terephthalate fibers and PET fibers which are welded to said particulate foam and a second portion of at least one of synthetic and natural reinforcing fibers comprising sufficient wettability for at least one of a rigidifying polymer, a hardening polymer and a linked



polymer forming a cover layer and disposed in a liquid phase onto a free surface of said mixed fiber layer.

74. The composite material of claim 73, wherein said polymer disposed onto said free surface of said mixed fiber layer is a thermoplastic material.
75. The composite material of claim 74, wherein said polymer disposed onto said free surface of said mixed fiber layer is at least one of polyalkylene terephthalate and PET.
76. The composite material of claim 73, wherein said polymer disposed onto said free surface of said mixed fiber layer is an elastomer.
77. The composite material of claim 73, wherein said polymer disposed onto said free surface of said mixed fiber layer is a thermosetting plastic material.
78. The composite material of claim 73, wherein said polymer disposed in its liquid phase onto said free surface of said mixed fiber layer is fiber-reinforced.
79. The composite material of claim 73, wherein said polymer disposed in said liquid phase onto said free surface of said mixed fiber layer comprises at least one of a

decorative layer, a foil, and a textile on a visible side thereof.

80. The composite material of claim 57, wherein said layer of material is a supporting layer which is one of disposed on a free surface of said particulate foam and disposed within said particulate foam.
81. The composite material of claim 80, wherein said supporting layer comprises at least one compact insertion part.
82. The composite material of claim 80, wherein said supporting layer comprises at least one polymer and is of compact structure.
83. The composite material of claim 82, wherein said supporting layer comprises at least one of polyalkylene terephthalate and PET.
84. The composite material of claim 80, wherein said supporting layer is welded to said particulate foam.
85. The composite material of claim 57, wherein the composite material has one of a multiple-layered structure and a sandwiched layer structure, wherein at least one composite material layer comprises a

2025041702

particulate foam of one of polyalkylene terephthalate and PET.

86. A molded part comprising the composite material of claim 57.
87. Use of the molded part of claim 86 for at least one of inner linings and technical structural parts in automotive vehicles.
88. Use of the molded part of claim 86 for at least one of furniture and garden furniture.
89. Use of the molded part of claim 86 for at least one of a sports device, a surf board, a wave slider, and a hull.
90. Use of the molded part of claim 86 for at least one of packings, insulation containers, and housings.
91. A method for producing a molded part from a composite material manufactured according to claim 57, wherein said particulate foam comprises at least one of polyalkylene terephthalate and pre-foamed polyalkylene terephthalate containing a foaming agent with a low crystallite portion in an otherwise amorphous phase, said particulate foam being heated in a mold to a temperature at which the surfaces of said particulate

foam melt and connect to one another to form said molded body, and, after cooling, at least one cover layer is laminated onto a free surface of said particulate foam molded body, wherein said particulate foam molded body is tempered through appropriate temperature control during at least one of production and cooling thereof.

92. The method of claim 91, wherein said particulate foam is brought to a temperature at which surfaces of said particulate foam melt in a presence of an intermediate layer containing polyalkylene terephthalate, said intermediate layer is back-foamed, a composite thereby obtained is cooled, and a cover layer is subsequently laminated onto said intermediate layer.
93. A method for producing a molded part from a composite material manufactured in accordance with claim 57, wherein at least one cover layer is heated in a mold together with a least one of densely packed particles of foamed polyalkylene terephthalate and pre-foamed polyalkylene terephthalate containing a foaming agent to a temperature at which at least a surface of said particles melts, said cover layer is back-foamed, and the composite material is subsequently cooled, wherein a resulting particulate foam molded body is tempered by appropriate temperature control during at least one of production and cooling.

94. The method of claim 93, further comprising disposing an intermediate layer between said particles and said cover layer, wherein said intermediate layer is welded to both said cover layer as well as to said particulate foam via back-foaming thereof.
95. A method for the production of a molded part from the composite material manufactured in accordance with claim 57, wherein an intermediate mixed fiber layer having a first portion of fibers of at least one of polyalkylene terephthalate and PET, and a second portion of reinforcing fibers having sufficient wettability for a liquid-phase polymer forming a cover layer, and densely packed particles of at least one of foamed polyalkylene terephthalate and pre-foamed polyalkylene terephthalate containing at least one foaming agent, are heated in a mold to a temperature at which surfaces of said particles and said polyalkylene terephthalate fibers of said mixed fiber layer melt, a resulting composite is subsequently cooled, and a liquid-phase polymer which is at least one of rigidifying, hardening and linking is disposed in a liquid phase onto a free surface of said mixed fiber layer, wherein said particulate foamed molded body is tempered through appropriate temperature control during at least one of production and cooling.

96. The method of claim 95, wherein said liquid-phase polymer is mixed with at least one of synthetic and natural reinforcing fibers and subsequently disposed onto said free surface of said mixed fiber layer.
97. The method of claim 95, further comprising disposing one of a decorative layer, a foil, and a textile onto a visible side of said polymer disposed in the liquid phase.
98. The method of claim 97, wherein said decorative layer is disposed onto said mixed fiber layer together with said liquid-phase polymer by one of injection and pressing.
99. The method of claim 91, wherein said particles are heated in said mold together with one of a compact support layer and a compact support layer containing at least one polymer, to a temperature at which at least surfaces of said particles melt, wherein foam is disposed behind or around said support layer.
100. The method of claim 91, wherein at least one of said particles, said cover layer, an intermediate layer, and a supporting layer are heated to the melting temperature by a diffusing hot gas phase.


101. The method of claim 91, wherein at least one of said particles, said cover layer, an intermediate layer and a supporting layer are heated to said melting temperature by means of microwave energy.

102. The method of claim 101, further comprising addition of a medium which absorbs microwaves.

#### REMARKS

The amendments have been taken to adapt this application to United States practice. No new matter has been added.

Lichti, Lempert and Lasch  
Bergwaldstr. 1  
D-76227 Karlsruhe, Germany  
Telephone: +49-721-9432815  
Fax: +49-721-9432840  
-9432850

  
\_\_\_\_\_  
Dr. Paul Vincent  
Agent for the Applicant  
Registration No. 37,461

4004632-041702

1/PRTS

19036.3

Translation of PCT/EP00/06881 as filed on July 19, 2000

Method for Producing a Composite Material, Composite Material  
Produced According to Said Method, Molded Part That Consists  
of Such a Composite Material and Method for the Production  
Thereof

The invention concerns a method for producing a composite  
material from a particulate foam that consists of a  
thermoplastic material and at least one layer connected  
thereto by heating the pre-foamed particles to a temperature  
in the region of the melting temperature and simultaneously  
or subsequently connecting to the layer. The invention also  
concerns a composite material produced with this method, that  
consists of a particulate foam and at least one layer  
connected thereto, molded parts made from such a composite  
material, and also methods of producing same.

Thermoplastic materials and plastic foams produced therefrom  
by expansion, especially particulate foams, are used for  
various applications. Plastic foams are mainly used as  
insulating heat, sound or impact layers and for reducing  
weight compared to compact materials. However, they absorb  
forces only to a limited degree and are usually neither  
abrasion-resistant nor diffusion-tight. In most applications,  
the plastic foam must therefore be coated at least on one



side or be totally covered, wherein the coating or covering material must provide a surface that is e.g. smooth, if necessary decorative, abrasion-resistant or sealing in dependence on the application, and/or should enhance the solidity of the molded part. Towards this end, the coating or covering material can e.g. be fiber-reinforced. Such composite materials are used in technical components, such as panel pieces in automobile technology, for housings, packings or the like.

After production and molding of the plastic foam of such composite materials, the plastic foam is e.g. connected to a cover layer by means of adhesives, solvents, etc.

Alternatively, the cover layer is prefabricated and subsequently back-foamed with the foam particles. The first method is expensive and usually harmful to the environment and stresses can lead to delaminations. If the plastic foam and the cover layer consist of different polymers, the connection between cover layer and plastic foam is often inadequate. The cover layer can delaminate or crack under excessive stress thereby disintegrating the plastic foam or even decomposing the foam structure.

Composite materials of polyurethane foams are known having coated, e.g. laminated or glued, decorative sheets of polyurethane or polyvinyl chloride (PVC). Due to their thermosetting plastic properties, polyurethanes can disadvantageously be recycled to only a limited degree and for that reason must be either thermally recycled or be

granulated using demanding and expensive methods. This granulated matter is then often used for the fabrication of low-quality products. If the decorative sheet consists of PVC, recycling is even more difficult because of the variety in grades. Thermal recycling is also problematic due to the chloride content. Thermal utilization of plastic materials containing PVC produces polychlorinated dibenzodioxins and furans. An example thereof is the 2,3,7,8-tetrachlorodibenzodioxin, known as "toxin of Seveso", that has the highest toxicity of all currently known organic compounds.

DE 37 22 873 describes a method for producing a piece of equipment, in particular a panel piece for automobiles, comprising a thin-walled support layer, an intermediate layer and a decorative layer. The support layer is melted to the plastic foam, and the decorative layer is pressed onto the plastic foam, preferably by flame lamination, or is coated in a conventional fashion. The plastic foam consists of polyethylene, and the support layer consists of sheet metal or of a fiber mat containing polypropylene, or of a fiber glass material containing polypropylene. Polyolefin foams have poor thermal shape stability, being stable in shape only up to approximately 100°C to 110°C. This is insufficient for many molded parts that are e.g. exposed to sunlight, hot air or exhaust heat. This is particularly true for the automobile industry which requires thermal shape stability up to a temperature of at least 120°C.

DE 41 41 113 A1 discloses a composite body in the form of an automobile panel piece that consists of a polyolefin particulate foam and a decorative layer, also consisting of polyolefin polymers, which is laminated thereon. The decorative layer has a multilayer knitted fabric or tissue with embedded spacers of polymer fibers and has a decorative textile surface. Alternatively, the decorative layer is coated with a laminated sheet based on olefin polymers. This composite material is pure in grade but also has poor thermal shape stability.

It is the underlying object of the invention to provide a method of producing an environmentally friendly composite material with a tight bond between the particulate foam and the layer that is linked therewith, and with a higher heat resistance and resistance to thermal shock. The object also includes such composite materials, molded parts from such a composite material and methods for producing such molded parts.

This object is achieved in accordance with the invention by using a particulate foam of polyalkylene terephthalate, or of a polyalkylene terephthalate blend, with a low crystallite portion in an otherwise amorphous phase, and tempering the particulate foam, during production of the molded body, during bonding with the layer and/or subsequently and at a temperature at which the amorphous phase is converted into a higher crystallite portion.

JP 20 73 836 A discloses introduction of foamed particles together with granulated matter, made from a polymer having a low melting point, into an extruder and controlling the temperature of the extruder such that only the granulated matter is melted. The blend of molten polymer and foamed particles is then transferred to an injection mold. The molded body consists of a polymer matrix with embedded foamed particles. PET is mentioned along with a plurality of other polymers. No selection criteria is given.

The invention makes use of the improved thermal stability and resistance to thermal shock of the polyalkylene terephthalates, in particular of PET, which have melting points in the region of 250°C and softening points in the region of 200°C. However, these relatively high temperatures for the production of the molded body from the particulate foam require a correspondingly high energy input. For the conventional production of molded bodies using superheated steam, a steam pressure of more than 16 bar would be needed. The required energy input is therefore considerable. The mold device must be correspondingly constructed to provide it with the required tightness under pressure, especially at the separation location. The molding tools must have a corresponding wall thickness which increases not only the investment costs but also the energy demand due to the high heat capacity of the mold. Practical tests have shown that approximately 95 % of the energy input is used for heating the mold. This prevents economic production of the molded parts.

According to the inventive method, the foamed or pre-foamed polyalkylene terephthalate is used in a configuration with low crystallite portion in an otherwise amorphous phase with the softening temperature of said polyalkylene terephthalate being considerably lower than the softening temperature of polyalkylene terephthalates in conventional applications. However, such a polyalkylene terephthalate of low crystallite portion does not have the material properties that are desirable for the finished molded body or molded part. It has turned out that the crystallite portion of the amorphous phase can be considerably increased by tempering the particulate foam molded body during or after production so that the particulate foam in the composite material exhibits the usual desired properties. In this fashion, the particles can be linked using less energy and at reduced investment cost. Conventional molding machines can also be used for the processing of particulate foams from polyalkylene terephthalates.

If the molded body is formed from the particulate foam with pressurized superheated steam, the pressure and the pressurizing time can be controlled for tempering the particulate foam.

It is also possible to continue tempering the composite material after production of the molded body, e.g. by slow and/or controlled cooling using the heat within the molded body. In particular, the composite material can be removed

from the mold immediately after having reached the necessary natural stability at the surface, and then slowly cooled.

The temperature control that is necessary for producing as high a crystallite portion as possible can be effected using a conventional DSC-measurement during the process or with prototypes.

A composite material from a molded body of a particulate foam with at least one layer linked to the molded body, that is produced according to the inventive method is characterized in that the particulate foam consists of a polyalkylene terephthalate or a polyalkylene terephthalate blend.

The polyalkylene terephthalate particulate foam of the composite material according to the invention has extraordinarily high tensile and shear strength. Compared to other particulate foams, in particular the most common polystyrene foams, it is characterized by its high restoring force which prevents permanent compression points. It has a higher thermal shape stability than polyolefin particulate foams and is stable up to at least 120°C. Polyalkylene terephthalates, such as polyethylene terephthalates (PET), polypropylene terephthalates, polybutylene terephthalates, poly(1,4-cyclohexane dimethylene terephthalate)s etc. are thermoplastic polymers of high solidity, rigidity and shape stability, with good sliding and wear properties as well as high chemical resistance. Due to the thermoplastic properties, the polyalkylene terephthalate particulate foam

10034333.011702

can be easily recycled and is therefore environmentally friendly. Even thermal disposal thereof does not produce toxic halogenated hydrocarbons.

The particulate foam consists preferentially of PET. Due to the partially crystalline structure of its molecular chains, PET has a particular high solidity and density ( $D \approx 1,38 \text{ g/cm}^3$ ) and a high melting point of approximately  $260^\circ\text{C}$ . The partially crystalline structures and hence the material properties of PET can be modified by copolymerization with higher terephthalic acid esters or e.g. isophthalic acid.

To increase the solidity and especially the viscosity of the composite material, the particulate foam can have synthetic and/or natural reinforcing fibers, such as glass, metal, carbon, aramide, wood, cellulose, hemp fibers etc. These reinforcing fibers are preferentially arranged between and within the particles.

The composite material according to the invention can be produced as sheets or as a three-dimensional molded part, if necessary also as a hollow body, e.g. by molding the composite material onto a core. The composite material according to the invention is therefore especially suited for molded articles for internal panels, engine compartments and car body parts, e.g. inner door panels, sun visors, consoles, dashboards, cockpit modules, linings of engine compartments, bumpers, hoods, front end supports, etc., for furniture, particularly garden furniture, sports equipment, such as

surfboards, wave boards, hulls, for packagings, insulation boxes or housings or for model construction.

Depending on the application of the finished molded part, the layer that is connected to the particulate foam is designed as a visible decorative layer which gives the composite material a smooth or structured, decorative surface and an improved splash, scratch and abrasion resistance. The cover layer can consist e.g. of at least one thermoplastic polymer, e.g. a polyolefin sheet to provide the composite body with a smooth surface. The cover layer can e.g. also have a textile structure, in order to give the composite body the desired optical and haptic features and e.g. a surface structure, in particular for inside panel pieces for automobiles.

In a preferred embodiment, the cover layer consists of polyalkylene terephthalate, especially PET, or a polyalkylene terephthalate blend. In this case, the composite material according to the invention can be completely recycled due to its grade purity and its thermoplastic properties and is therefore environmentally friendly. On the other hand, the particulate foam and the cover layer form a solid and durable connection due to the compatibility of identical or similar polymers.

In accordance with a preferred embodiment variant, the cover layer is welded to the particulate foam or is laminated onto the particulate foam, e.g. flame laminated, calendered, coated, etc. As already mentioned, the cover layer can be



formed by a deep drawn sheet consisting of polyalkylene terephthalate.

In a preferred version, the cover layer is applied onto the particulate foam via an intermediate layer of fibers, wherein the intermediate layer preferably has a fiber mat. The intermediate layer can also be a tissue, knitted fabric, cloth, knits or the like.

Preferentially, the intermediate layer consists of polyalkylene terephthalate, in particular PET, or a polyalkylene terephthalate blend. To increase the strength of the composite material, the intermediate layer can be reinforced with synthetic and/or natural reinforcing fibers, such as glass, carbon, aramide, metal, cellulose, wood, hemp fibers etc.

The intermediate layer that is preferentially welded to the particulate foam, does not thermally affect the particulate foam during lamination of the cover layer. On the other hand, subsequent foaming of an intermediate layer that is preferably welded to the cover layer, prevents the particle structure of the particulate foam from becoming visible by pressing through the cover layer. Furthermore, such an intermediate layer produces comfortable and soft haptic features for the molded part made from the composite material which is desirable for many applications.

According to another preferred variant of the embodiment, the intermediate layer is a mixed fiber layer which contains a portion of polyalkylene terephthalate fibers, in particular PET, which are welded to the particulate foam, and a further portion of synthetic and/or natural reinforcing fibers which have sufficient wettability for at least one solidifying, hardening and/or linking polymer which forms the cover layer and is disposed in the liquid phase onto the free surface of the mixed fiber layer. The intermediate layer thereby serves two functions within the composite material: the conventional reinforcing function with respect to solidity and viscosity and also the bonding between the cover layer and the polyalkylene terephthalate particulate foam, since it contains polyalkylene terephthalate fibers which form a close welding connection therewith when melting the particulate foam. This produces a strong bond between the intermediate, mixed fiber layer and the particulate foam. The free surface of the intermediate layer also has sufficient wettability for applying or impregnating a solidifying hardening or linking polymer such that a coating having the required properties, e.g. smooth surface, abrasion resistance, resistance to splashing and scratches etc., is disposed on the composite of mixed fiber layer and polyalkylene terephthalate particulate foam. The coating of a solidifying, hardening or linking polymer which is disposed on the free surface of the mixed fiber layer can, for its part, comprise several layers, e.g. have a sandwich-like design.

The polymer which has been disposed in the liquid phase onto the free surface of the mixed fiber layer can e.g. be a thermoplastic material disposed on the mixed fiber layer using any conventional method, such as injection molding or low-pressure injection molding, pressing on, extrusion or coextrusion, thermoplastic foam casting, thermal molding, flame injection, extrusion deposition or extrusion flow methods. The thermoplastic material, e.g. polyolefines, polystyrene, polyacrylates etc., which is transferred into the liquid molten phase by heating, solidifies during cooling on the mixed fiber layer, wherein this layer serves not only as a bonding agent and a reinforcement but, in particular, also as a heat insulation layer which thermally protects the particulate foam during injection, extrusion or the like.

The thermoplastic material polyalkylene terephthalate, in particular PET, disposed in the liquid phase is preferably such that the composite material is characterized by good thermal stability and high purity of grades as well as low material and production costs.

The polymer disposed in the liquid phase can also be a linking polymer in the form of an elastomer, e.g. caoutchouc. An elastomer provides additional functions in the surface region, such as improved grip, dampening of pressure and impact forces.

It is also possible to dispose a linking polymer in the form of a thermosetting plastic material onto the free surface of

the mixed fiber layer. The thermosetting plastic material can be disposed - like the elastomer - in any fashion, e.g. by slush techniques, soaking, impregnation, spraying or reaction injection molding (RIM or RRIM=reinforced reaction injection molding or SRIM=structural reaction injection molding) which is based on rapid admixing and blending of the liquid thermosetting components, injection of the reactive mixture onto the mixed fiber layer and rapid hardening. Liquid monomers or oligomers with integrated powdery polymers can be used, wherein these act like crystallization seeds during hardening or linking of monomers or oligomers on the mixed fiber layer, wherein the hardened and/or linked polymer has a low polymerization loss due to the portion of already polymerized particles. When thermosetting plastic materials or elastomers are used, the mixed fiber layer also acts as a heat insulation layer in order not to impair the particulate foam during the usually exothermal linking. Essentially all known thermosetting plastic materials can be used, e.g. polyurethane, epoxy, melamine, urea, formaldehyde or phenolic resins and compounds of these resins.

To improve the strength of the inventive composite material, the polymer which has been disposed in the liquid phase onto the free surface of the mixed fiber layer, can be reinforced with synthetic and/or natural fibers.

The polymer disposed in the liquid phase onto the free surface of the mixed fiber layer can also comprise a decorative layer, e.g. a sheet, textile or the like, at least

on the visible side, which can be disposed in any conventional fashion onto the solidified, hardened and/or linked polymer or at the same time when the liquid polymer is disposed onto the mixed fiber layer. The decorative layer can e.g. also be a sheet of veneer, e.g. wooden veneer which is glued onto the smooth surface of the solidified, hardened and/or linked polymer.

In a further development of the inventive composite material, at least one supporting layer is disposed on at least one free surface of the particulate foam or inside the particulate foam. The supporting layer preferably comprises at least one compact insertion part, e.g. a supporting element in the form of an injection molded part. It preferably consists of a compact polymer and also preferably contains polyalkylene terephthalate, in particular PET or is completely formed thereof. The supporting layer is preferably welded to the particulate foam. In this case, the supporting layer which consists of compact material imparts the largest possible stability and durability to the composite material, while the particulate foam serves e.g. as an impact, sound or heat insulation layer and reduces the weight of the composite material. If the composite material comprises a cover layer, as mentioned above, this cover layer provides a smooth or structured surface and improved resistance to scratching and abrasion.

The invention also concerns molded parts which consist essentially of a composite material of the above-mentioned

structure, and methods for producing such molded parts. An embodiment of such a method provides that densely packed particles of polyalkylene terephthalate having a low crystallite portion which is foamed and/or prefoamed and provided with a foaming agent, are heated in a mold to a temperature at which the surface of the particles melt, and connected into the molded body, and after cooling at least one layer, e.g. a cover layer, is laminated onto at least one free surface of the particulate foam molded body, and the particulate foam molded body is tempered during its production and/or during cooling through corresponding temperature control. The term "mold" designates herein any molding measure producing plate-shaped, spatial or hollow molded parts.

The densely packed polyalkylene terephthalate particles are heated to a temperature at which only the surface of the particles melts and interconnects. After cooling of the foamed body, the smooth or textiled cover layer, which consists in particular of thermoplastic polymers, e.g. polyalkylene terephthalate, is laminated by means of any technique, e.g. by flame lamination, deep drawing or the like. In a preferred embodiment, the particles are heated in the presence of a textile intermediate layer, e.g. a fleece of polyalkylene terephthalate, at which the surface of the particles melts, and the intermediate layer is foamed, the composite obtained is cooled and the cover layer is then laminated onto the intermediate layer. This ensures that during lamination of the cover layer, the particulate foam is

not thermally impaired and e.g. if the cover layer is a thin sheet, the foamed particles are not visible through the cover layer. This intermediate layer also produces soft haptic features.

In another method variant for producing a molded part from such a composite material at least one layer, e.g. a cover layer, and densely packed particles of foamed polyalkylene terephthalate or pre-foamed polyalkylene terephthalate provided with a foaming agent are heated in a mold to a temperature at which at least the surface of the particles melts, the cover layer is back-foamed, and the composite material is subsequently cooled. In this case as well, an intermediate layer, e.g. a fleece of polyalkylene terephthalate can be preferably disposed between the particles and the cover layer, and the intermediate layer can be preferably welded to both the cover layer as well as to the particulate foam during subsequent foaming thereof. The foaming produces a lasting connection among the layers wherein the one-step method guarantees rapid and inexpensive production of the inventive composite material. Alternatively, the intermediate layer can be initially connected to the cover layer, e.g. by welding with subsequent back-foaming of the intermediate layer.

In another method for producing a molded part from an inventive composite material, an intermediate layer in the form of a mixed fiber layer containing a portion of polyalkylene terephthalate fibers, in particular PET, and a

10034233-011702

further portion of reinforcing fibers having sufficient wettability for a polymer in liquid phase forming the cover layer, and densely packed particles of foamed polyalkylene terephthalate and/or polyalkylene terephthalate having at least one foaming agent which disintegrates in the region of the melting temperature of the polyalkylene terephthalate, are heated in a mold to a temperature at which the surface of the particles and the polyalkylene terephthalate fibers of the mixed fiber layer melt, the obtained composite is subsequently cooled and the solidified, hardened and/or linked polymer is disposed in the liquid phase onto the free surface of the mixed fiber layer. Since the intermediate, mixed fiber layer contains polyalkylene terephthalate fibers, these fibers are melted thereby producing a strong material link between the mixed fiber layer and the particulate foam. As mentioned above, a molten thermoplastic material or a hardening or linking polymer (elastomer or thermosetting plastic material) is subsequently disposed in the liquid phase onto the mixed fiber layer using any conventional technique and the desired molded part is obtained after solidification, hardening, or linking. The solidifying, linking or hardening polymer can e.g. connect two molded parts.

Alternatively, the cover layer can be prefabricated, e.g. the mixed fiber layer can be impregnated with the solidifying, hardening and/or linking polymer. Care should be taken that the polymer does not protrude through to the other side of the mixed fiber layer. The foamed particles are then disposed



onto the free surface of the mixed fiber layer and heated thereby simultaneously producing a strong connection to the polyalkylene terephthalate fibers of the mixed fiber layer.

In one embodiment, the solidifying, hardening and/or linking polymer is mixed in the liquid phase with synthetic and or natural reinforcing fibers and subsequently disposed onto the free surface of the mixed fiber layer. If the polymer is e.g. a thermoplastic material which is extruded onto the mixed fiber layer, the reinforcing fibers can be added to an extruder in the region of the extruder screw at which the thermoplastic material is substantially completely plastified. The reinforcing fibers can be added to the liquid polymer in any fashion irrespective of whether the polymer is a thermoplastic material, an elastomer or a thermosetting plastic material.

A decorative layer, e.g. a sheet, textile or the like can be optionally disposed on the visible side of the polymer disposed in the liquid phase. In a preferred embodiment, the decorative layer is simultaneously disposed onto the mixed fiber layer together with the polymer by injection, compression or the like. In this fashion, the polymer can be disposed in the liquid phase onto the composite consisting of particulate foam and mixed fiber layer in that the composite and the decorative layer are disposed in a mold and the liquid polymer which is optionally reinforced by fibers is introduced between composite and decorative layer using any conventional method. The smooth surface of the solidified,

hardened or linked polymer can then be easily provided with a decorative layer, e.g. having a glued-on wooden veneer.

All above methods permit heating the particles in the mold in the presence of a compact supporting layer to a temperature at which at least the surface of the particles melts, and foaming up of the support. The supporting layer preferably consists of a polymer or a polymer blend, e.g. of substantially compact polyalkylene terephthalate, such as PET.

The particles and optionally the cover layer and/or the intermediate layer can be heated to the melting temperature by means of a gas phase diffusing same, e.g. hot vapor. The diffusion-open structure of the layers leads to an intimate bonding of the particulate foam as well as among the layers.

Alternatively, the particles and optionally the cover layer and/or the intermediate layer and/or the supporting layer can be heated to the melting temperature using microwave energy, wherein a medium which absorbs microwaves, such as water, alcohol or the like, can preferably be used. The microwave-absorbing medium can be disposed e.g. in the liquid phase onto the surfaces to be connected and be removed from the mold after evaporation. In this fashion, the type and amount of the microwave-absorbing medium used produces a reliable welding connection and prevents local overheating in the mold.

Embodiments of the invention are explained in detail below with reference to the drawing.

Fig. 1 shows a cross-section of a molded part having the shape of a sports device produced from a composite material in accordance with the invention;

Fig. 2 shows a cross-section of a molded part in the shape of a lining part for the interior of automotive vehicles produced from a composite material in accordance with the invention.

The molded part 5a shown in Fig. 1, e.g. a surf board, comprises a compact support layer 1 which can consist e.g. of PET or of a blend of PET and other polyalkylene terephthalates. A particulate foam 2 surrounds the support layer 1 which e.g. also consists essentially of PET or of a blend of PET and other polyalkylene terephthalates and which is preferably welded to the support layer to produce a tight connection between the support layer 1 and particulate foam 2. An intermediate layer 4, i.e. a layer of mixed fibers, is disposed on the particulate foam having a portion of polyalkylene terephthalate fibers which are welded to the particulate foam 2. The mixed fiber layer also comprises reinforcing fibers, e.g. glass or carbon fibers which have sufficient wettability for a rigidified, hardened, or linked polymer, e.g. polyurethane, disposed in the liquid phase onto the free surface of the mixed fiber layer to form a cover layer 3. The cover layer 3 provides high diffusion tightness,

resistance to spraying, scratching and abrasion. It can also have a sandwich-like design of several superposed layers.

Fig. 2 shows a molded part 5b of uniform material, e.g. a dashboard 7 of an automotive vehicle including instrument housing 6. The molded part 5b consists e.g. of a PET particulate foam 2 and a decorative cover layer 3 of PET disposed on its visible side. The cover layer 3 can be e.g. in the form of a textile, such as a tissue, knitted fabric or the like. An intermediate layer 4 disposed between particulate foam 2 and cover layer 3, which consists e.g. of a PET fleece gives the molded part 5b soft haptic features. If the cover layer 3 is laminated onto the particulate foam 2, the intermediate layer 4 provides additional thermal protection for the particulate foam 2 when applying the cover layer 3. If the intermediate layer 4, which is e.g. welded to the cover layer 3, is subsequently back-foamed with the particulate foam 2, penetration of the particulate foam 2 through the visible surface of the cover layer 3 is prevented.

## Claims

1. Method for producing a composite material from a particulate foam of thermoplastic material, and at least one layer connected thereto by heating the pre-foamed particles to a temperature in the region of the melting temperature and connecting them together to form a molded body and simultaneously or subsequently connecting to the layer, characterized in that a particulate foam of polyalkylene terephthalate, or a blend of polyalkylene terephthalate, having a low crystallite portion in an otherwise amorphous phase is used, and that the particulate foam is tempered, when forming the molded body, when forming the composite with the layer, and/or subsequent thereto, at a temperature converting the amorphous phase into an increased overall crystallite portion.
2. Method according to claim 1, characterized in that the particulate foam is formed into the molded body using pressurized hot vapor and the pressure or the pressurizing time for tempering the particulate foam is controlled.
3. Method according to claim 1 or 2, characterized in that the molded body is further tempered after production.
4. Method according to any one of the claims 1 through 3, characterized in that the molded body is slowly cooled after production.

5. Method according to any one of the claims 1 through 4, characterized in that the temperature during tempering is controlled by a DSC measurement.
6. Composite material, consisting of a molded body of a particulate foam and at least one layer connected to the molded body, produced in accordance with the method according to any one of the claims 1 through 5, characterized in that the particulate foam (2) consists of a polyalkylene terephthalate or a blend of polyalkylene terephthalates.
7. Composite material according to claim 6, characterized in that the particulate foam (2) consists of polyethylene terephthalate (PET).
8. Composite material according to claim 6 or 7, characterized in that the particulate foam comprises synthetic and/or natural reinforcing fibers.
9. Composite material according to any one of the claims 6 through 8, characterized in that the layer connected to the particulate foam (2) is a cover layer (3) forming the visible side.
10. Composite material according to claim 9, characterized in that the cover layer (3) is a foil of at least one thermoplastic polymer.

11. Composite material according to claim 9, characterized in that the cover layer (3) comprises a textile structure.
12. Composite material according to any one of the claims 9 through 11, characterized in that the cover layer (3) consists of polyalkylene terephthalate, in particular PET, or a blend of polyalkylene terephthalates.
13. Composite material according to any one of the claims 9 through 12, characterized in that the cover layer (3) is welded to the particulate foam (2).
14. Composite material according to any one of the claims 9 through 12, characterized in that the cover layer (3) is coated onto the particulate foam (2).
15. Composite material according to any one of the claims 6 through 14, characterized in that an intermediate layer (4) of fibers is disposed between the cover layer (3) and the particulate foam (2).
16. Composite material according to claim 15, characterized in that the intermediate layer (4) is a fiber mat, cloth, tissue, knitted fabric or knits.
17. Composite material according to claim 16 or 17, characterized in that the fibers of the intermediate layer

consist of polyalkylene terephthalate, in particular PET, or a polyalkylene terephthalate blend.

18. Composite material according to any one of the claims 15 through 17, characterized in that the intermediate layer (4) comprises synthetic and/or natural reinforcing fibers.
19. Composite material according to any one of the claims 15 through 18, characterized in that the intermediate layer (4) is welded to the particulate foam (2).
20. Composite material according to any one of the claims 15 through 19, characterized in that the intermediate layer (4) is welded to the cover layer (3).
21. Composite material according to any one of the claims 15 through 20, characterized in that the cover layer (3) is laminated onto the intermediate layer (4).
22. Composite material according to any one of the claims 15 through 21, characterized in that the intermediate layer (4) is a mixed fiber layer containing a portion of fibers of polyalkylene terephthalate, in particular PET, which are welded to the particulate foam (1) and a further portion of synthetic and/or natural reinforcing fibers comprising sufficient wettability for at least one rigidifying, hardening and/or linked polymer which forms the cover layer (3) and which is disposed in the liquid phase onto the free surface of the mixed fiber layer.



23. Composite material according to claim 22, characterized in that the polymer disposed onto the free surface of the mixed fiber layer is a thermoplastic material.
24. Composite material according to claim 23, characterized in that the polymer disposed onto the free surface of the mixed fiber layer is polyalkylene terephthalate, in particular PET.
25. Composite material according to claim 22, characterized in that the polymer disposed onto the free surface of the mixed fiber layer is an elastomer.
26. Composite material according to claim 22, characterized in that the polymer disposed onto the free surface of the mixed fiber layer is a thermosetting plastic material.
27. Composite material according to any one of the claims 22 through 26, characterized in that the polymer disposed in its liquid phase onto the free surface of the mixed fiber layer is fiber-reinforced.
28. Composite material according to any one of the claims 22 through 27, characterized in that the polymer disposed in the liquid phase onto the free surface of the mixed fiber layer comprises, at least on its visible side, a decorative layer, e.g. a foil, textile or the like.

29. Composite material according to any one of the claims 6 through 28, characterized in that at least one supporting layer (1) is disposed on a free surface of the particulate foam (2) or within the particulate foam (2).
30. Composite material according to claim 29, characterized in that the supporting layer comprises at least one compact insertion part.
31. Composite material according to claim 29 or 30, characterized in that the supporting layer (1) consists of at least one polymer and is compact.
32. Composite material according to claim 31, characterized in that the supporting layer (1) comprises polyalkylene terephthalate, in particular PET, or is formed exclusively thereof.
33. Composite material according to any one of the claims 28 to 32, characterized in that in that the supporting layer (1) is welded to the particulate foam (2).
34. Composite material according to any one of the claims 6 through 33, characterized in that several layers are provided, in particular in the form of sandwiches, at least one of which is a particulate foam (2) of polyalkylene terephthalate, in particular PET.

35. Molded part (5a,5b) of a composite material in accordance with any one of the claims 6 through 33.
36. The use of a molded part in accordance with claim 35 for inner linings or as technical structural parts for automotive vehicles.
37. The use of a molded part in accordance with claim 35 for furniture, in particular garden furniture.
38. The use of a molded part in accordance with claim 35 for sports devices, such as surf boards, wave sliders, hulls or the like.
39. The use of a molded part in accordance with claim 35 for packings, insulation containers or housings.
40. Method for producing a molded part (5a,5b) from a composite material according to any one of the claims 6 through 34, characterized in that particles of foamed polyalkylene terephthalate, and/or pre-foamed polyalkylene terephthalate containing a foaming agent, with a low crystallite portion in an otherwise amorphous phase, are heated in a mold to a temperature at which the surface of the particles melts and connect to one another to form the molded body, and after cooling, at least one layer, e.g. a cover layer (3) is laminated onto a free surface of the particulate foam molded body, and that the particulate

foam molded body is tempered through appropriate temperature control during production and/or cooling.

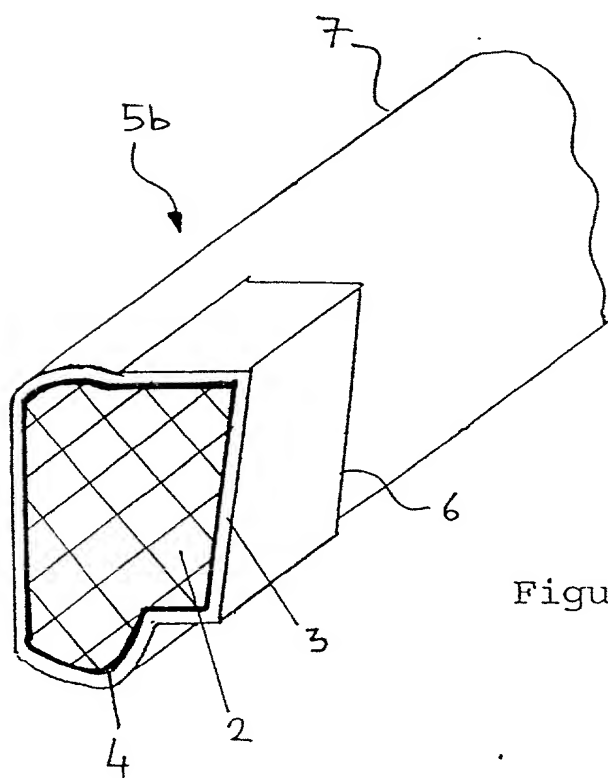
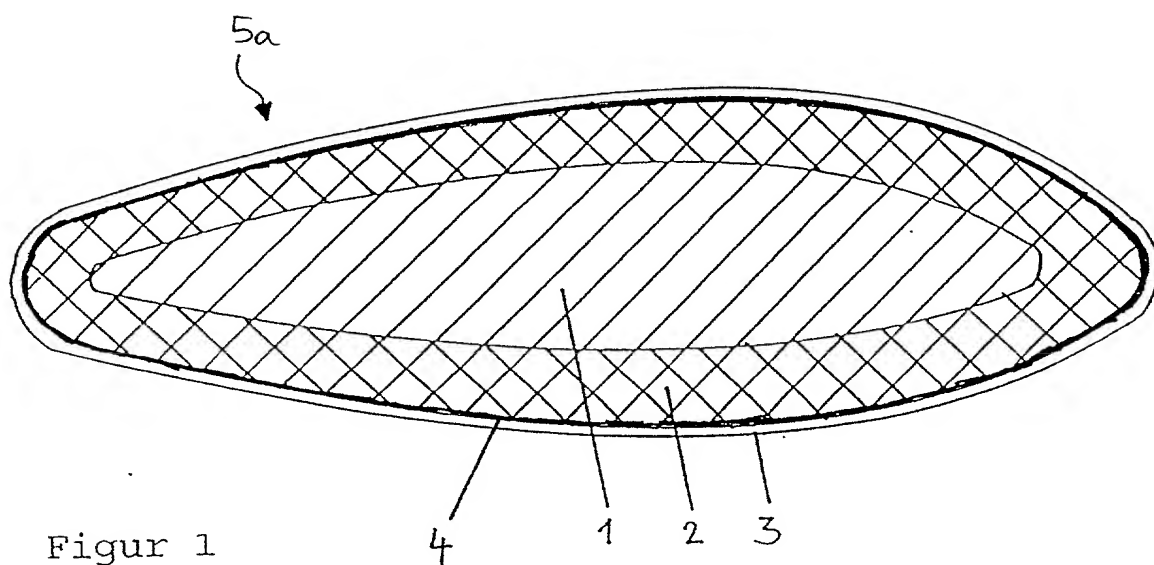
41. Method according to claim 40, characterized in that the particles are brought to a temperature at which the surface of the particles melts in the presence of an intermediate layer (4) containing polyalkylene terephthalate, and the intermediate layer (4) is back-foamed, the composite obtained is cooled and the cover layer (3) is subsequently laminated onto the intermediate layer (4).
42. Method for producing a molded part (5a,5b) from a composite material in accordance with any one of the claims 6 through 34, characterized in that at least one layer, e.g. a cover layer, and densely packed particles of foamed, and/or pre-foamed polyalkylene terephthalate containing a foaming agent, are heated in a mold to a temperature at which at least the surface of the particles melts, the cover layer (3) is back-foamed, the composite material is subsequently cooled, and the particulate foam molded body is tempered by appropriate temperature control during production and/or during cooling.
43. Method according to claim 42, characterized in that an intermediate layer (4) is disposed between the particles and the cover layer (3) and the intermediate layer (4) is welded to both the cover layer (3) as well as to the particulate foam (2) via back-foaming thereof.

44. Method for the production of a molded part from a composite material in accordance with any one of the claims 5 through 34, characterized in that an intermediate layer (4) in the form of a mixed fiber layer (2) having a portion of fibers of polyalkylene terephthalate, in particular PET, and a further portion of reinforcing fibers having sufficient wettability for a liquid-phase polymer forming the cover layer, and densely packed particles of foamed polyalkylene terephthalate, and/or pre-foamed polyalkylene terephthalate containing at least one foaming agent, are heated in a mold to a temperature at which the surface of the particles and the polyalkylene terephthalate fibers of the mixed fiber layer melt, the obtained composite is subsequently cooled and then, the rigidifying, hardening and/or linked liquid-phase polymer in the liquid phase is disposed onto the free surface of the mixed fiber layer, wherein the particulate foamed molded body is tempered through appropriate temperature control during its production and/or during cooling.
45. Method according to claim 44, characterized in that the rigidifying, hardening and/or linked liquid-phase polymer is mixed with synthetic and/or natural reinforcing fibers and subsequently disposed onto the free surface of the mixed fiber layer.

46. Method according to claim 44 and 45, characterized in that a decorative layer, e.g. a foil, textile or the like is disposed onto the visible side of the polymer disposed in the liquid phase.
47. Method according to claim 46, characterized in that the decorative layer is disposed onto the mixed fiber layer together with the polymer by injection, pressing or the like.
48. Method according to any one of the claims 40 through 47, characterized in that the particles are heated in the mold in the presence of a compact support layer (1), in particular, of at least one polymer, to a temperature at which at least the surface of the particles melts, wherein foam is disposed behind or around the supporting layer (1).
49. Method according to any one of the claims 40 through 48, characterized in that the particles and optionally the cover layer (3) and/or the intermediate layer (4) are heated to the melting temperature by a hot gas phase which diffuses through them.
50. Method according to any one of the claims 40 through 48, characterized in that the particles and optionally the cover layer (3) and/or the intermediate layer (4) and/or the supporting layer (1) are heated to the melting temperature by means of microwave energy.

51. Method according to claim 50, characterized in that a medium is used which absorbs microwaves.

COPIED FROM MICROFILM





## COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

(Includes Reference to PCT International Applications)

ATTORNEY DOCKET NUMBER

19036.3

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

METHOD FOR PRODUCING <sup>A</sup>COMPOSITE MATERIAL, COMPOSITE MATERIAL  
~~PRODUCED ACCORDING TO SAID MATERIAL, MOLDED PART THAT CONSISTS~~  
OF SUCH A COMPOSITE MATERIAL AND METHOD FOR THE PRODUCTION THEREOF

the specification of which (check only one item below):

☐ is attached hereto.

☐ was filed as United States application

Serial No. \_\_\_\_\_

on \_\_\_\_\_

and was amended

on \_\_\_\_\_ (if applicable)

☒ was filed as PCT international application

Number PCT/EP00/06881

on July 19, 2000

and was amended under PCT Article 19

on \_\_\_\_\_ (if applicable)

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is known to me or other person(s) involved in the preparation or prosecution of this application to be material to the examination of this application and to patentability as defined in Title 37, Code of Federal Regulations, §1.56

I hereby authorize the U.S. attorney or agent named herein to accept and follow instructions from \_\_\_\_\_

as to any action taken in the Patent and Trademark Office regarding this application without direct communication between the U.S. attorney or agent and the undersigned. In the event of a change in the persons from whom instructions may be taken, the U.S. attorney or agent named herein will be so notified by the undersigned.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

## PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119

COUNTRY (if PCT, indicate "PCT")	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 USC 119	
Germany	199 34 692.5	23. July 1999	<input checked="" type="checkbox"/> YES	<input type="checkbox"/> NO
Germany	100 11 331.1	10. March 2000	<input checked="" type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO
			<input type="checkbox"/> YES	<input type="checkbox"/> NO

<b>Combined Declaration For Patent Application and Power of Attorney (Continued)</b> <small>(Includes Reference to PCT International Applications)</small>				<b>ATTORNEY DOCKET NUMBER</b>	
<p>I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:</p>					
<b>PRIOR U.S. APPLICATIONS OR PCT INTERNATIONAL APPLICATIONS DESIGNATING THE U.S. BENEFIT UNDER 35 U.S.C. 120</b>					
<b>U.S. APPLICATIONS</b>			<b>STATUS (Check one)</b>		
U.S. APPLICATION NUMBER	U.S. FILING DATE	PATENTED	PENDING	ABANDONED	
PCT APPLICATIONS DESIGNATING THE U.S.					
PCT APPLICATION NUMBER	PCT FILING DATE	U.S. SERIAL NUMBERS ASSIGNED (if any)			
<p><b>POWER OF ATTORNEY:</b> As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (List name and registration number)</p> <p style="text-align: center; margin-top: 20px;"> <u>Paul J. Vincent</u>    Reg. No <u>37.461</u> </p>					
Send Correspondence to: <div style="margin-left: 20px;"> <u>Lichti, Lempert, &amp; Lasch</u>  <u>Bergwaldstr. 1</u>  <u>D-76227 Karlsruhe, Germany</u> </div>			Telephone <u>49-721-943 28 15</u> Fax <u>49-721-943 28 40</u> <u>+ 943 28 50</u>		
201	FULL NAME OF INVENTOR <u>Ziegler</u>	FAMILY NAME <u>Ziegler</u>	FIRST GIVEN NAME <u>Maik</u>	SECOND GIVEN NAME	
201	RESIDENCE & CITIZENSHIP <u>Pfinztal</u>	CITY <u>Pfinztal</u>	STATE OR FOREIGN COUNTRY <u>Germany DEX</u>	COUNTRY OF CITIZENSHIP <u>Germany</u>	
201	POST OFFICE ADDRESS <u>Am Hohberg 8</u>	POST OFFICE ADDRESS <u>Am Hohberg 8</u>	CITY <u>D-76327 Pfinztal</u>	STATE & ZIP CODE/COUNTRY <u>Germany</u>	
202	FULL NAME OF INVENTOR <u>Hofmann</u>	FAMILY NAME <u>Hofmann</u>	FIRST GIVEN NAME <u>Knut</u>	SECOND GIVEN NAME	
202	RESIDENCE & CITIZENSHIP <u>Bretten</u>	CITY <u>Bretten</u>	STATE OR FOREIGN COUNTRY <u>Germany DEX</u>	COUNTRY OF CITIZENSHIP <u>Germany</u>	
202	POST OFFICE ADDRESS <u>Händelstr. 8</u>	POST OFFICE ADDRESS <u>Händelstr. 8</u>	CITY <u>D-75015 Bretten</u>	STATE & ZIP CODE/COUNTRY <u>Germany</u>	
203	FULL NAME OF INVENTOR 	FAMILY NAME 	FIRST GIVEN NAME 	SECOND GIVEN NAME 	
203	RESIDENCE & CITIZENSHIP 	CITY 	STATE OR FOREIGN COUNTRY 	COUNTRY OF CITIZENSHIP 	
203	POST OFFICE ADDRESS 	POST OFFICE ADDRESS 	CITY 	STATE & ZIP CODE/COUNTRY 	
<p>I hereby declare under penalty of perjury under the laws of the United States of America that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon</p>					
SIGNATURE OF INVENTOR 201 		SIGNATURE OF INVENTOR 202 		SIGNATURE OF INVENTOR 203 	
DATE <u>17.12.01</u>		DATE <u>17-12-2001</u>		DATE 	